APPLICATION NOTE

Los Alamos National Laboratory

Passive/Active Neutron Coincidence Counter

April 1993

Safeguards Assay Group N-1

MS E540

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LALP 93-31

This work was supported by the US Department of Energy, Office of Safeguards and Security.

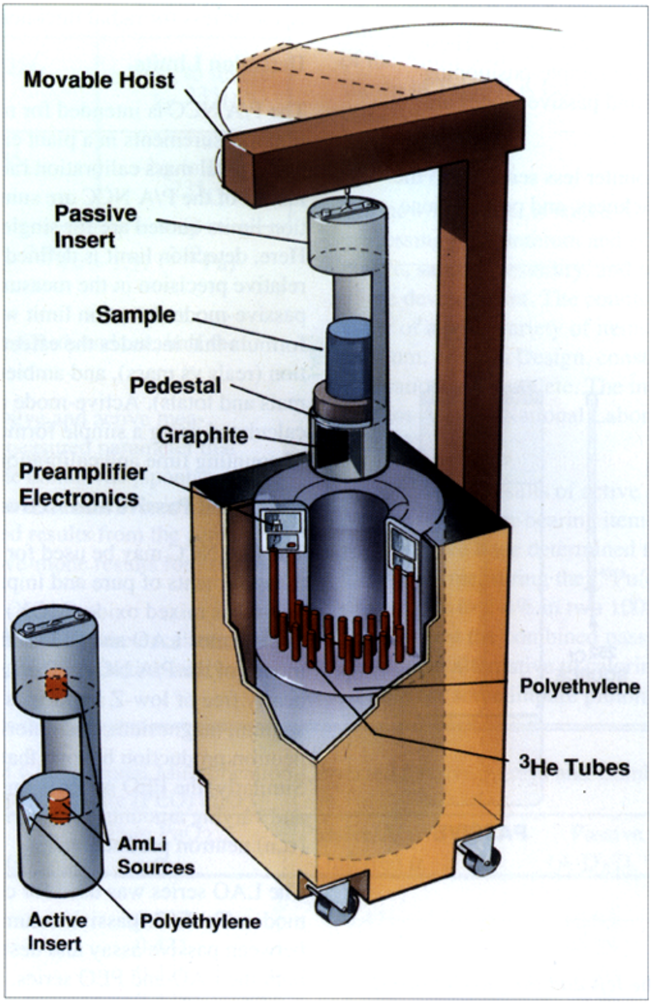

Fig. 1. In the passive/active well counter shown at left, a plutonium sample (silver-grey) rests on a pedestal (silver) inside the cavity of the passive insert. Graphite in the top and bottom cylinders of the passive insert reflects fast neutrons from the sample, thereby improving counting efficiency and radiation shielding. The movable hoist (brown) lowers the insert into the well. The active, fast mode insert is shown at far left. Here AmLi neutron sources are located in the upper and lower polyethylene cylinders. After several collisions, AmLi neutrons induce fissions in uranium samples. Fission neutrons emitted by the sample (either uranium or plutonium) slow down in the polyethylene moderator and are then detected by the ^{3}He tubes (red) that surround the well. Pulse-processing electronics modules (inside the junction box above the ^{3}He tubes) produce digital neutron pulses for subsequent time-correlation processing and counting circuits. The insert-lifting mechanism has several unique features including both up- and down-limit switches and a quickly actuating motor brake for safe, flexible, and efficient sample handling.

The new passive/active well-type neutron coincidence counter (P/A NCC) is designed for nondestructive assay of a wide variety of items containing plutonium, uranium, or both. Plant personnel can make both active and passive measurements with one instrument by simply changing the well inserts. By combining the results of active and passive measurements (measurements with the active insert but with and without the AmLi sources), operators can assay a wider variety of impure plutonium-bearing items. The neutron moderator body is an annulus of polyethylene containing forty-eight ^{3}He proportional counters in two concentric rings. The ^{3}He-tube placement and moderator design were optimized to produce a measured efficiency of 37.4% with the passive insert. The active inserts have a measured efficiency of 32.7% in the fast mode and 37.0% in the thermal mode. The efficient design yields sensitive measurements for both uranium and plutonium. The counter can produce passive measurements of plutonium-bearing samples, active thermal-mode measurements of samples with low ^{235}U masses (<50 g ^{235}U), and active fast-mode measurements of samples with high ^{235}U masses (>50 g ^{235}U).

Performance Goals

One of the primary performance goals for the P/A NCC was a nearly constant axial efficiency profile for both passive and active modes. The multiplication-corrected reals efficiency is uniform to within ±1% from 50 to 350 mm (~2-14 in.) above the cavity floor. In the active-fast mode, response is lowest in the cavity center, but is uniform to within ±5% for samples with fill-heights between 2.5 and 20 cm. In the active-thermal mode, response is uniform to within ±1% for samples with fill-heights between 2.5 and 20 cm. These profiles yield negligible biases from sample-positioning effects. The inserts for the active and passive modes are shown in Fig. 2.

It was also desired to make the counter less sensitive to the sample matrix, steel container thickness, and polyethylene covering bags. The new counter is approximately a factor of 2 less sensitive to the matrices sand, ash, lead shot, and iron shot than similar counters. In passive mode, we measured separately the effects of steel and polyethylene shells surrounding the sample. The effect of external steel on the multiplication-corrected reals rate is essentially nil for radial thicknesses between 0 and 5 mm. The effect of the external polyethylene on the multiplication-corrected reals is 1% or less for radial thicknesses between 0 and 12 mm.

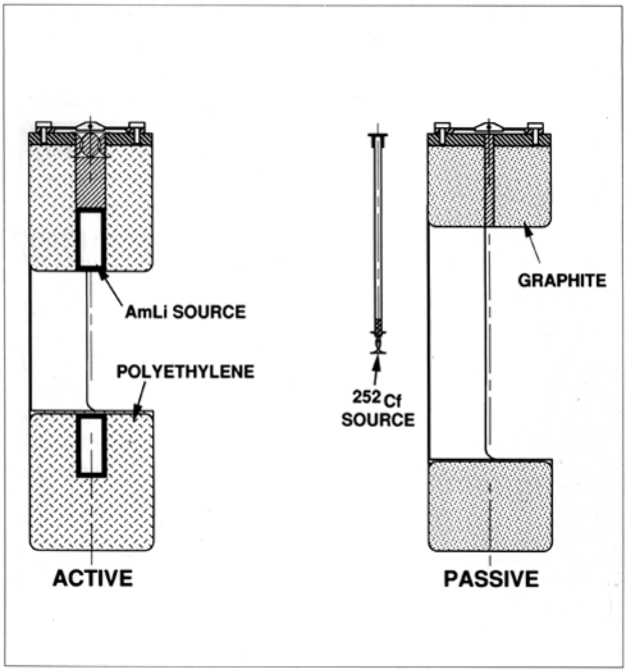

Fig. 2. The active insert is on the left and the passive insert on the right. The ^{252}Cf source holder is shown with the passive insert. The passive insert contains graphite end plugs (each 178 mm or 7 in. thick) for optimum reflection (and minimal absorption) of neutrons originating in the plutonium-bearing sample. Also, the top plug allows placement of a ^{252}Cf source in the center of the sample cavity for routine measurement-control checks. The passive cavity will hold sample containers up to 216 mm (8.5 in.) in diameter and 406 mm (16 in.) high. The active inserts hold polyethylene end plugs (each 273 mm or 10.75 in. thick) that include receptacles for AmLi sources immediately above and below the sample cavity. The first active insert has cadmium surrounding the sample cavity (fast mode). The second insert has no cadmium (thermal mode). The active cavities will accommodate sample containers up to 216 mm (8.5 in.) in diameter and 229 mm (9 in.) tall.

Detection Limits

The P/A NCC is intended for routine assay and/or verification measurements in a plant environment. Detection limits and typical mass calibration ranges for the three operating modes of the P/A NCC are summarized in Table I. Detection limits quoted are for single 1000-s measurements. Here, detection limit is defined as the mass value yielding a relative precision in the measured reals rate of 33%. The passive-mode detection limit was calculated using a general formula that includes the effects of counting time, calibration (reals vs mass), and ambient background rates (both reals and totals). Active-mode detection limits were calculated using a simple formula that includes the effects of counting time and calibration (reals vs mass).

Combined Passive and Active Measurements

The P/A NCC may be used for combined active and passive measurements of pure and impure plutonium-bearing items as well as mixed oxide (MOX) samples. We measured the Los Alamos LAO and PEO series of PuO_2 standards in all modes of the P/A NCC. The LAO series is known to be nearly free of low-Z impurities (for example, fluorine, sodium, magnesium, and chlorine) that increase the (α,n) neutron production beyond that expected from pure PuO_2. Similarly, the PEO series is known to contain significant and varying amounts of impurities and therefore excess (α,n) neutron production.

The LAO series was used for calibration in all measurement modes. Table II (passive column) shows the differences between passive assay and destructive analysis values for both the LAO and PEO series. Because the LAO series was used for calibration, the LAO assay results are excellent; the average difference is +0.12 ± 0.63%. However, for the PEO series, the average difference is +8.83 ± 3.51% because we used a value for α [ratio of (α,n) neutrons to spontaneous fission neutrons] that is calculated for pure PuO_2 and therefore too low for the impure samples. Passive measurement times varied between 600 and 2000 s. Using simple propagation of the errors in the totals and reals, the resulting precision in the multiplication-corrected reals (R_c) ranged between 0.05 and 0.1%. We concluded that 0.25% precision in R_c can be obtained in only a 100-s counting time for passive-mode measurements of items similar to the LAO and PEO standards. Compared to the standard High-Level Neutron Coincidence Counter (HLNCC-II) that has an efficiency of 17.5% and the Flat-Squared Counter with an efficiency of 24.4%, the P/A NCC can obtain the same

TABLE I. Passive/Active Counter Detection Limits and Calibration Ranges		
Measurement Mode	1000-s Measurement Detection Limits	Mass Calibration Range
Passive	1–2 mg $^{240}Pu_{eff}$*	~5 mg to a few kg $^{240}Pu_{eff}$
Active-Fast	50 g ^{235}U	~100 g to several kg ^{235}U
Active-Thermal	1 g ^{235}U	~2 g to 100 g ^{235}U

*$^{240}Pu_{eff} = 2.49\ (^{238}Pu) + {}^{240}Pu + 1.57\ (^{242}Pu)$

passive counting precision in one-fourth and one-half the counting times, respectively.

By combining results of both passive and active measurements, we obtained another measured parameter that allows solving for all three sample unknowns [spontaneous fission neutrons, (α,n) neutrons, and induced-fission or multiplication neutrons]. We used results from the active measurement to correct the passive-mode results for sample multiplication.

For the impure PEO standards, the results are excellent; the average difference between assay and destructive-analysis values is +0.08 ± 0.80%. Measurement times for the passive measurements varied between 600 and 1800 s, with one measurement of 5400 s for the smallest sample. Measurement times for the active measurements were much longer. These varied between 1000 and 50 000 s, with two measurements of 100 000 s for small samples. Precisions were also calculated for two 1000-s measurements: one active and one passive. We estimate that for two 1000-s measurements, the scatter in the assay-declared differences would be 3-4%. This accuracy could be improved by increasing the strengths of the AmLi sources. Most of the scatter in the assay values shown in Table II is from causes other than counting statistics.

Summary

The new P/A NCC is suited for general use in plants processing both uranium and plutonium. Optimum performance, safety, versatility, and reliability were primary goals of the development. The counter provides nondestructive assay of a wide variety of items containing plutonium, uranium, or both. Design, construction, testing, and initial evaluation are complete. The instrument has been installed at the Los Alamos National Laboratory Plutonium Facility (TA-55).

By combining results of active and passive measurements, impure plutonium-bearing items and MOX samples can be assayed. We have determined that this new approach is capable of measuring the ^{240}Pu(eff) mass in impure plutonium oxide to 2-4% in two 1000-s measurements. Initial data suggest the combined passive/active approach is a reasonable alternative to calorimetry or multiplicity counting or both for some impure plutonium-bearing items.

TABLE II. Comparison of Conventional Multiplication-Corrected Passive Assays and Combined Passive/Active Assays of Pure (LAO) and Impure (PEO) PuO_2

Sample	$^{240}Pu_{eff}$ (g)	Pure PuO_2 α	Passive/Active inferred α	Passive (A-D)/D, %	Passive/Active (A-D)/D, %
LAO1	10.098	0.427	0.471	+0.93	-0.12
LAO2	29.296	0.431	0.472	+0.28	-3.37
LAO3	54.359	0.426	0.463	+0.70	-2.81
LAO4	65.115	0.425	0.408	-0.88	+0.40
LAO5	92.188	0.433	0.399	-0.63	+2.46
LAO6	104.537	0.426	0.401	+0.49	+2.62
LAO7	144.365	0.423	0.411	-0.06	+1.07
LAO8	149.126	0.423	0.414	+0.12	+0.85
			Average	+0.12	+0.14
			Std. Dev.	0.63	2.21
PEO1	7.418	0.622	0.870	+12.54	-0.38
PEO2	14.836	0.622	0.817	+11.75	-0.88
PEO3	29.672	0.622	0.788	+11.14	-0.02
PEO4	43.339	0.625	0.704	+6.71	+1.50
PEO5	64.998	0.615	0.722	+7.20	-0.04
PEO6	81.374	0.622	0.677	+3.66	+0.28
			Average	+8.83	+0.08
			Std. Dev.	3.51	0.80

TABLE III. Advantages of the New Passive/Active Neutron Coincidence Counter	
Feature	**Advantage**
Removable sample-well inserts and AmLi sources	Allow separate passive and active measurements
Fast- and thermal-mode inserts for active measurements	Allow uranium assay in both low- and high-mass ranges
Large sample cavities	Accommodate a wide range of containers
Intense AmLi sources	Override passive coincidence signals for mixed uranium and plutonium, impure plutonium samples, or both
Radiation shielding	Reduces effects of background neutrons and reduces personnel exposure to neutrons and gamma rays
Uniform efficiency in the sample cavity	Minimizes sample positioning effects
Low sensitivity to sample matrix, containers, and coverings	Reduces variability in response caused by sample variability
Passive efficiency of 37.4%	Excellent counting efficiency
Active efficiency of 37.0% in thermal mode and 32.7% in fast mode	Excellent counting efficiency

Additional Sources of Information

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